Estimation of the optical constants and thickness of thin films using unconstrained optimization

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1 Introduction

The transmission of a thin film on a transparent substrate is a function of the wavelength (λ) and given by:[1]

$$T(\lambda) = \frac{Ax}{B - Cx + Dx^2}$$

where A, B, C, and D are functions of $n(\lambda)$, $k(\lambda)$, and d − the refractive index, the extinction coefficient, and the thickness of the film, respectively − and the refractive index of the substrate, assumed to be known. We wish to estimate d, $n(\lambda)$ and $k(\lambda)$ from a given set of experimental data $[\lambda_i, T_{meas}(\lambda_i)]$, $i = 1, \ldots, N$, ($\lambda_{min} \leq \lambda_i < \lambda_{i+1} \leq \lambda_{max}$ for all $i = 1, \ldots, N$). For any d and λ, the equation: $T_{calc}[\lambda, s(\lambda), d, n(\lambda), k(\lambda)] = T_{meas}(\lambda)$ must hold. Unfortunately, the equation has two unknowns $n(\lambda)$ and $k(\lambda)$, and the problem is highly underdetermined. Its set of solutions is, in general, a curve in the two-dimensional $[n(\lambda), k(\lambda)]$ space. Therefore, the set of functions (n, k) satisfying it for a given d is infinite and, roughly speaking, is represented by a nonlinear manifold of dimension N in $\mathbb{R}^{2N}$.

The index of refraction and the thickness of transparent films displaying a fringe pattern can be estimated with a reasonable accuracy.[2] Assuming a functional dependence for $n(\lambda)$, the method may be extended to estimate the extinction coefficient in the absorbing spectral region.[3] In general, these approximations are of limited accuracy. In a recent publication we considered the problem from a different point of view.[4] Instead of imposing functional forms, the physical constraints that $n(\lambda)$ and $k(\lambda)$ must satisfy in the measured spectral range was explicitly established. These constraints represent a prior knowledge about the solution. The estimation problem takes the form:

$$\text{Minimize } \sum_{\lambda} [T_{calc}(\lambda) - T_{meas}(\lambda)]^2$$

subject to the appropriate physical constraints. In this way, well behaved functions $n(\lambda)$ and $k(\lambda)$ can be obtained without severe restrictions that could damage the quality of the retrieval. The method has been successfully applied to all kind of transmission spectra and does not rely on the existence of fringe patterns or transparency. The main inconvenient of the above pointwise constrained optimization approach is that it is a rather complex large-scale linearly constrained nonlinear programming problem whose solution can be obtained only by means of rather sophisticated and not always available computer codes that can deal effectively with sparsity of the matrix of constraints.[5,6]

The main contribution of the present paper is to establish a method for solving the estimation problem replacing the above constrained by an *unconstrained* optimization problem. We solved this problem using a very simple algorithm introduced recently by Raydan.[7] This method realizes a very effective idea for potentially large-scale unconstrained minimization. It consists of using only gradient directions with step-lengths that ensure a rapid convergence. The reduction of a constrained to an unconstrained minimization problem needed the calculation of complicate derivatives of functions, which could not be possible without the use of automatic differentiation techniques. Here we used the procedures for automatic differentiation described in Ref. 8.

2 Unconstrained formulation of the estimation problem

The spectral region $[\lambda_{\min}, \lambda_{\max}]$ covered by optical transmission of thin dielectric films goes from complete opacity to transparency, or to some degree of transparency, and corresponds to normal dispersion. In this spectral region the refractive index n is a convex function of λ, whereas the absorption coefficient $\alpha(\lambda) = \frac{4\pi k(\lambda)}{\lambda}$ is generally convex at small absorption and concave at large absorption.

The pointwise optimization approach introduced in Ref. 4 is as follows: for each tentative thickness value d the sum of squares $\sum_i [T_{calc}(\lambda_i, n_i, \alpha_i) - T_{meas}(\lambda_i)]^2$ is minimized subject to the constraints:

1. $n(\lambda) \geq 1$ and $\alpha(\lambda) \geq 0$ for all $\lambda \in [\lambda_{\min}, \lambda_{\max}]$,

2. $n(\lambda)$ and $\alpha(\lambda)$ are decreasing functions of λ,

3. $n(\lambda)$ is convex,

4. there exists $\lambda_{infl} \in [\lambda_{\min}, \lambda_{\max}]$ such that $\alpha(\lambda)$ is convex if $\lambda \geq \lambda_{infl}$ and concave if $\lambda < \lambda_{infl}$.

This gives a nonlinear programming problem with linear constraints that was solved using a well established software.

In this work, we observe that given a set of experimental points λ_i ($i = 1, \ldots, N$) the constraints of the problem are equivalent to:

- $n_N = 1 + v^2$, $\alpha_N = u_N^2$,
- $n_{(N-1)} - n_N = v_{(N-1)}^2$, $\alpha_{(N-1)} - \alpha_N = u_{(N-1)}^2$,
- $n_{(i+2)} - 2n_{(i+1)} + n_i = v_i^2$, for $i = 1, \ldots, N-2$,
- $\alpha_{(i+2)} - 2\alpha_{(i+1)} + \alpha_i = -u_i^2$, for $i = 1, \ldots, i_{infl}$,
- $\alpha_{(i+2)} - 2\alpha_{(i+1)} + \alpha_i = u_i^2$, for $i = i_{(infl+1)}, \ldots, N-2$,

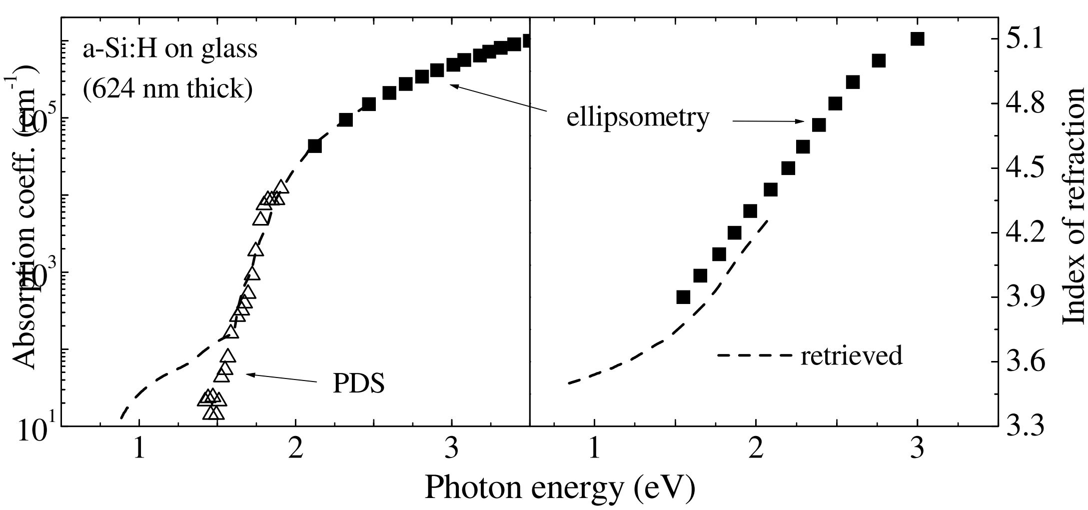
where $u_1, \ldots, u_N, v_1, \ldots, v_N$ are auxiliary variables. Clearly, we can isolate $\alpha_1, \ldots, \alpha_N, n_1, \ldots, n_N$ as functions of $u_1, \ldots, u_N, v_1, \ldots, v_N$, so that the optimization problem is now:

$$\boxed{\text{Minimize } \sum_i [T_{calc}(\lambda_i, u_i, v_i) - T_{meas}(\lambda_i)]^2}$$

without constraints at all, since these are automatically satisfied by the change of variables. (In fact we work here with the extinction coefficient k_i instead of α_i, but this does not affect the essence of the new formulation). The unconstrained approach instead of the constrained one adds simplicity to our codes and portability of the whole software package.

3 Numerical applications

The new method was applied to the solution of a variety of thin film spectra.[9] As in Ref. 4 the reliability of the approach was tested with computer generated films, the solution of which was known in advance. In all cases the



retrieval process was satisfactory, giving the same accuracy as that found with the constrained optimization approach.[4] For practical purposes, however, the calculation of real films is of prime importance. As an example, we present here the optical properties retrieved for an a-Si:H film deposited onto Corning 7059 glass, which were also determined from independent methods.[4] See the figure below.

Note the three orders of magnitude retrieval of the absorption coefficient and its good agreement with the values determined from ellipsometry and photothermal deflection spectroscopy (PDS) on the same film. The index of refraction was also retrieved with a reasonable accuracy. The thickness giving the best fit to measured transmission data (624 nm) is the same as that measured with a profilometer and also to that inferred from interference fringes. It is important to remember here that the goodness of the fit does not depend on the existence of a fringe pattern or on film transparency.

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[9] The detailed treatment will appear in a forthcoming publication.